FABRICATION AND CHARACTERIZATION OF ATOMIZED U-MO POWDER DISPERSED FUEL COMPACTS FOR THE RERTR-3 IRRADIATION TEST

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ABSTRACT

The RERTR-3 irradiation test of nano-plates will be conducted in the Advanced Test Reactor(ATR). 49 compacts with a uranium density of 8 gU/cc consist of 7 different atomized uranium-molybdenum alloy powders such as as-atomized U-10Mo, phase decomposed U-10Mo (alpha+gamma), homogenized U-10Mo, U-7Mo, U-6Mo, U-6.1Mo-0.9Ru, and U-6Mo-1.7Os. 25 fuel plates, referred to as nano-plates, were produced with atomized fuel compacts at ANL-W. The relationship between the volume fraction of fuel and the green density of the compacts was established. The relative density of the compacts increases with decreasing volume fraction of fuel powder. The compressibility of comminuted powder compacts was larger than that of the atomized powder compacts due to the fragmentation of comminuted particles. The green strength of comminuted powder compacts is higher than that of the atomized powder compact. This seems to have resulted from the smaller pore size and the larger contact area between the comminuted fuel powders and Al powders. It is suggested that the compacting condition adjustment be required to fabricate the atomized powder compacts having comparable green strength.

I. INTRODUCTION

The goal of recent fuel development in RERTR program is to produce a new low enriched uranium (LEU) fuel meat with a uranium density of 8 to 9gU/cm^3 [1]. There have been two methods to accomplish this goal. One is the increase of the uranium content in the fuel alloy and the other is the increase of the amount of fuel dispersant in the fuel meat. Efforts to increase the amount of fuel dispersant in the fuel meat were already made by CERCA. CERCA's results showed that the maximum fuel loading in Almatrix was about 55 vol%[2]. Consequently, new fuel materials should have a uranium density of about 15 gU/cm^3 to fabricate the fuel meat with a uranium density of 8 to 9gU/cm^3.

Therefore, gamma-stabilized U-Mo alloys have been suggested as new fuel material[1].

At KAERI, the centrifugal atomization process has been applied to fabricate fuel powders since about 10 years ago. U-Mo alloy powders as well as uranium silicide powders which are spherical in shape have been successfully fabricated[3]. Because the U-Mo alloys have a ductile nature, the alloys cannot be converted to powder form by the processes routinely used for oxides or intermetallics[4]. Therefore, the atomization is well suited as a unique commercial process for the fabrication of U-Mo alloy powder to meet RERTR program needs[4]. Moreover, the experimental results of two recent irradiation tests designated RERTR-1 and RERTR-2, performed in the Advanced Test Reactor(ATR), showed that the atomized U-10Mo alloy exhibited excellent irradiation performance[5]. The new RERTR irradiation test designated RERTR-3 is scheduled to be performed in ATR. A total of 48 experimental fuel plates, referred to as nano-plates, will be irradiated in ATR from October 1999[6]. The 25 nano-plates fabricated with atomized U-Mo alloy compacts which were supplied by KAERI.

The green strength of a cold pressed powder compact is an important property, since most industrial processes necessitate a certain amount of handling[7]. Especially, fuel compacts with optimum green strength are required to produce the plate type fuel meat. The results of BWXT showed that the atomized U_3Si_2 spherical particles were more segregated than the comminuted particles in fuel meat and the number of stray particles in atomized U_3Si_2 fuel meat was larger than that in comminuted U_3Si_2 fuel meat[8]. The inhomogeneity and the evolution of stray particles in atomized fuel meat is closely related to the green strength of the compact.

In this study, the fabrication process and specification of atomized U-Mo alloy compacts for nano-plate are presented. 49 compacts with a uranium density of 8 gU/cc consist of 7 different atomized uranium-molybdenum alloy powders such as as-atomized U-10Mo, phase decomposed U-10Mo (alpha+gamma), homogenized U-10Mo, U-7Mo, U-6Mo, U-6.1Mo-0.9Ru, and U-6Mo-1.7Os. The relationship between the volume fraction of fuel and the green density of the compacts was established. In addition, the green density and green strength were determined for the compacts of U-10Mo/Al and U_3Si_2/Al at a compacting pressure ranging from 50 to 400 MPa. The effects of the volume fraction and the shape of the fuel powder on green properties were investigated and the optimum fabrication conditions of compacts suitable for producing the fuel plate using atomized powder were established.

II. PRELIMINARY TEST OF POWDER COMPACTS

The specifications of powder compacts for nano-plates, which ANL sent to KAERI, are as shown in Table 1. Because the powder compacts are fabricated from very a small amount of powder mixture(below 0.7g), it is difficult to obtain compacts having a nominal volume fraction of fuel powders. Moreover, it is impossible to know not only the actual volume fraction of U-10Mo powders but also the relative density in the mixtures. Thus, a preliminary test was performed to fabricate the power compacts having exactly 52 vol% U-10Mo powders. In order to accomplish this goal, several power mixtures with a total amount of 0.7g, including 0.61g of U-10Mo and 0.09g of Al, were mixed by hand and were compacted at the pressure range of 210-506 MPa. Although the homogeneity of these mixtures is very poor, the volume fractions of the hand-mixed powders lie at the exact value of 52 vol %. The optimum compacting pressure of 380 MPa was fixed to obtain the powder compact having a relative density of above 95% and to satisfy the RERTR fabrication procedure[9]. Table 1 shows the properties of hand-mixed power compacts.

As shown in Table 2, it was confirmed that all the hand-mixed compacts exhibited the compacts with relative densities above 95%(below 5% of porosity). In order to confirm our experimental results, the U-10Mo/Al compacts with 60 and 70 vol % fuel powder by hand-mixing were fabricated at the pressure of 380 MPa. Relative densities of 96.3% for 60 vol% powder mixture and 93.3% for 70 vol% powder mixture were obtained. Therefore, it was confirmed that the relative densities of 52 vol% U-10Mo/Al compacts were above 95% at the pressure of 380MPa.

Fig. 1 shows the relationship between the volume fraction of U-10Mo and the measured density of the compacts having above 95% relative density. This figure shows that the compacts with relative density above 95% have densities of 9.0 to 10g/cm^3. The volume fraction of U-10Mo in compacts are between 45%(for 9g/cm^3 of measured density and 100% of relative density) and 55%(for 10g/cm^3 of measured density and 90% of relative density).

III. FABRICATION OF POWDER COMPACTS FOR NANO-PLATES

The powders of U-10Mo, U-7Mo, U-6Mo, U-6.1Mo-0.9Ru, and U-6Mo-1.7Os were prepared by centrifugal atomization method. Powders less than 120 mesh in size were obtained by sieving. Decomposed U-10Mo(alpha+gamma) powder was prepared by heat treating atomized U-10Mo powder at 500^{o}C for 500hr and gamma homogenized U-10Mo powder was prepared after heat treatment of as-atomized U-10Mo powder at 800^{o}C for 100hr. The compositions of fuel alloys were designed by KAERI and ANL.

Fig. 2 shows the cross-sectional SEM micrographs of as-atomized U-10Mo powder, decomposed U-10Mo powder and homogenized U-10Mo powder. The as-atomized U-10Mo powder exhibits a cell structure with less than 5μm in cell size as shown in Fig. 2(a). Kim et al[10] reported that the atomized U-10Mo powder exhibited the cell structure due to a Mo micro-segregation at grain boundaries. The compositional analysis of atomized U-10Mo powder by EDS showed that the Mo content at the cell boundary is about 2-3 at% lower than that in the cell interior[11]. Several previous studies also reported that the atomized powder had cell structure with the cell size of 10-100μm [12] and the solute content of cell boundary was smaller than that of cell interior[13].

The micrograph of decomposed U-10Mo powder as shown in Fig. 2(b) shows that most γ cells are decomposed into α-U and γ′(U_2Mo) phases by eutectoid decomposition. It has been reported that the decomposition of γ phase at 500°C took place by the so called cellular mechanism[11]. The nucleation initiated at cell boundaries having lower Mo content and γ′ phase, precedently precipitated near the cell boundaries by the conventional eutectoid decomposition. The SEM micrograph of homogenized U-10Mo powder as shown in Fig. 2(c) exhibits a featureless microstructure. The homogenized U-10Mo particle consists of a single grain due to coarsening of cell structures. Fig. 3 shows the cross-sectional SEM micrographs of as-atomized U-6Mo powder, U-7Mo powder, U-6.1Mo-0.9Ru powder and U-6.05Mo-1.7Os powder. All as-atomized powders shown in Fig. 3(a)-(d) exhibited the cell structure with the cell size of below 5μm, similar to the as-atomized U-10Mo powder shown in Fig 2(a).

Table 3 shows the description of 49 fuel powder compacts, including the fuel composition, alloy U-density, density of compacts, relative density of compacts and fuel loading. The U-density of all the compacts is about $8gU/cm^3$. U-Mo alloy powder and pure Al powder were blended using off-axis drum mixer with various fuel volume fractions as shown in Table 3. The blended powder was compacted to a pellet having nominal dimension of 4.0mm diameter by 4.8mm height at a pressure of 380 MPa. The pressure die for the fabricating compact was supplied by ANL. Pressure was held for 10 seconds and no lubricant was used to prevent contamination. The weight and dimension of compacts were measured and the relative density of each compact was calculated. All fabricated meeting requirement were sent to ANL. The error range of volume fraction of the fuel in compact could be calculated based on Fig. 2. The measured volume fractions of compacts are shown in Table 3 with the error range of ±2%. All the compacts were successfully rolled into nano-plate and all nano-plates fabricated with atomized powders appeared to be satisfactory.

IV. GREEN DENSITY OF POWDER COMPACT

The green properties of powder compacts of atomized U-10Mo, atomized U_3Si_2 and comminuted U_3Si_2 powders were investigated. U-10Mo/Al and U_3Si_2/Al powders were blended using an off-axis drum mixer with various volume fractions of fuel powder. The blended powders were compacted to powder compacts having nominal dimension of 12.8mm wide by 31.8mm long by 6.4mm thickness at a pressure range of 50-400 MPa. Pressure was held for 10 second. Since the green properties of the compacts are not affected by a small amount of surface contamination, 10% zinc strearate in ethanol was used as lubricant for convenient fabrication. Table 4 summarizes the fabrication conditions of the compacts for green strength measurement. The green densities of the compacts were calculated from the weights and the dimensions. The green strengths of compacts were determined according to ASTM standard[14].

Fig. 4 shows the relative densities of compacts of pure Al, 40 vol%, 50 vol% and 60 vol% atomized U-10Mo/Al powders with varying pressure. The relative density increased both with increasing pressure and with decreasing volume fraction of U-10Mo powder at pressure up to 300 MPa. The compacts of 60vol% atomized U-10Mo powder and pure Al did not densify more at pressure above 350 MPa, while the densities of compacts of 40 vol% and 50 vol% atomized U-10Mo continuously increased at the pressure up to 400 MPa. Fig. 5 represents the relative densities of various compacts of 50 vol% atomized U-10Mo, 50 vol% atomized U_3Si_2 and 50 vol% comminuted U_3Si_2 powders with various pressures. The compressibility of the compact of 50 vol% atomized U-10Mo was larger than that of the compact of 50 vol% U_3Si_2 at pressure above 200 MPa. The relative densities of the compacts of comminuted U_3Si_2 were larger than those of the compacts of atomized U_3Si_2.

Fig. 6 shows SEM micrographs of the compacts pressed at the pressure of 400 MPa. Deformation of U-10Mo powder after compaction was observed as shown in Fig. 6(a)-(c). Deformation of U-10Mo powder is suggested as a main reason for lager compressibility of the compact of U-10Mo compared to the compact of U_3Si_2 at pressure above 200 MPa. The microstructure of the compact of atomized U_3Si_2 showed cracks in the particles, while the microstructure of the compact of comminuted U_3Si_2 exhibited fragmentation of fuel particles as shown in Fig. 6(d) and 6(e). A small number of large pores adjacent to the atomized U_3Si_2 particle, and a large number of small pores adjacent to the comminuted U_3Si_2 particle were observed. It has been reported that the compressibility of spherical metallic powder was larger than that of irregular shaped metallic powder[15]. However, in case of the compacts of U_3Si_2, the compressibility of the comminuted U_3Si_2/Al powder was founded to be larger than that of atomized U_3Si_2/Al powder due to the particle fragmentation during compaction.

V. GREEN STRENGTH OF POWDER COMPACTS

Fig. 7 shows the green strengths of powder compacts of 40 vol%, 50 vol% and 60 vol% atomized U-10Mo with varying pressures. Green strengths of compacts increased with increasing pressure and with decreasing the volume fraction of U-10Mo powder. Fig. 8 shows the green strengths of compacts of 50 vol% atomized U_3Si_2, comminuted U_3Si_2 and atomized U-10Mo with various pressures. The green strength of comminuted compacts was almost twice as large as than that of the atomized compact as shown in Fig. 8. The green strength of a compact is mainly due to the bonding force between the interparticle metallic contacts during the compacting process[16] and is proportional to the contact area between the particles[17]. However, most of the previous studies on the green strength of compacts focused on compacts made by mono-sized spherical metallic powder[16-18]. In this study, we used irregular shaped aluminum powder(ALCAN 101) and spherical and comminuted fuel powders having a somewhat broad size distribution. Therefore, it is impossible to apply the quantitative dependence of green strength on the contact area, derived from geometrical factors in previous studies.

Fig. 9 shows green strengths of the compacts of 40 vol%, 50 vol% and 60 vol% atomized U-10Mo with varying relative density. Fig. 10 shows the green strengths of the compacts of 50 vol% atomized U_3Si_2, comminuted U_3Si_2 and atomized U-10Mo with relative density. The green strength of the compacts of atomized U-10Mo exhibited similar dependence on the relative density regardless of their volume fraction of fuel powder. The green strengths of the compacts of comminuted U_3Si_2 were much larger than those of the compacts of atomized powder. The green strength of a powder compact is mainly due to the bonding force between the interparticle metallic contact during the compacting process and bonding force between the interparticle metallic contact enhances as the plastic deformation of particles increases[19]. The bonding force between Al particles is much larger than that between Al and fuel particles and therefore the green strength of the compact is controlled by the contact area between Al and fuel particle. The contact area between Al and comminuted fuel particles was larger than that between Al and atomized fuel particles. Therefore the green strength of comminuted fuel/Al compact is much larger than that of the atomized fuel/Al compact. It is certain that the particle shape plays a major role affecting the green strength of Al/fuel compacts.

The RERTR fabrication procedure for plate-type fuel meat indicated that the relative density of 90-

95% was optimum for the compacts of U_3Si_2. The green strengths of the compact of 50 vol% comminuted U_3Si_2 having a relative density of 91-93%, are about 12-18MPa as shown in Fig. 10. Therefore it is concluded that the compacts of atomized U-10Mo having a relative density above 97% are required in order to fabricate atomized fuel compacts having comparable green strength with comminuted fuel compacts.

VI. CONCLUSIONS

1. Sound compacts for nano-plates meeting the required specification were successfully fabricated.
2. The compressibility of comminuted powder compacts was larger than that of the atomized powder compacts due to the fragmentation of comminuted particles. The compressibility of the compacts of U-10Mo was larger than that of the compacts of U_3Si_2 due to the deformation of U-10Mo particles.
3. The green strength of the comminuted powder compacts was about twice as large as that of the atomized powder compacts, since the contact area between Al and comminuted fuel particles was larger than that between Al and atomized fuel particles
4. The compacts of atomized U-10Mo having a relative density above 97% are required in order to fabricate atomized fuel compacts having comparable green strength with comminuted fuel compacts.

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Table 1. Specification of Powder Compact for Nano-plate

	Height(mm)	Diameter(mm)*	Powder Size	Relative Density(%)**
Range	3.96±0.08	4.81	-120 mesh	90-95

* Originally 4.76mm, modified. ** Acceptable up to full density

Table 2. Properties of Hand-mixed Powder Compacts

Number of Mixture	Pressure (MPa)	Compact Weight (g)	Density of Compact (g/)	Relative Density (%)
1	380	0.6940	9.94	99.1
2	380	0.6860	9.47	94.4
3	380	0.6935	9.76	97.3

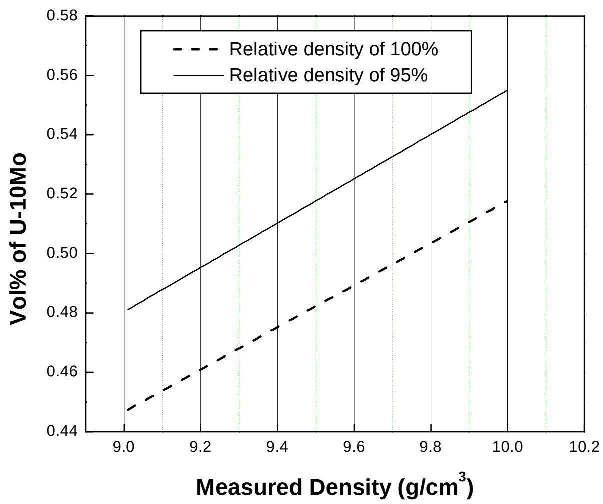
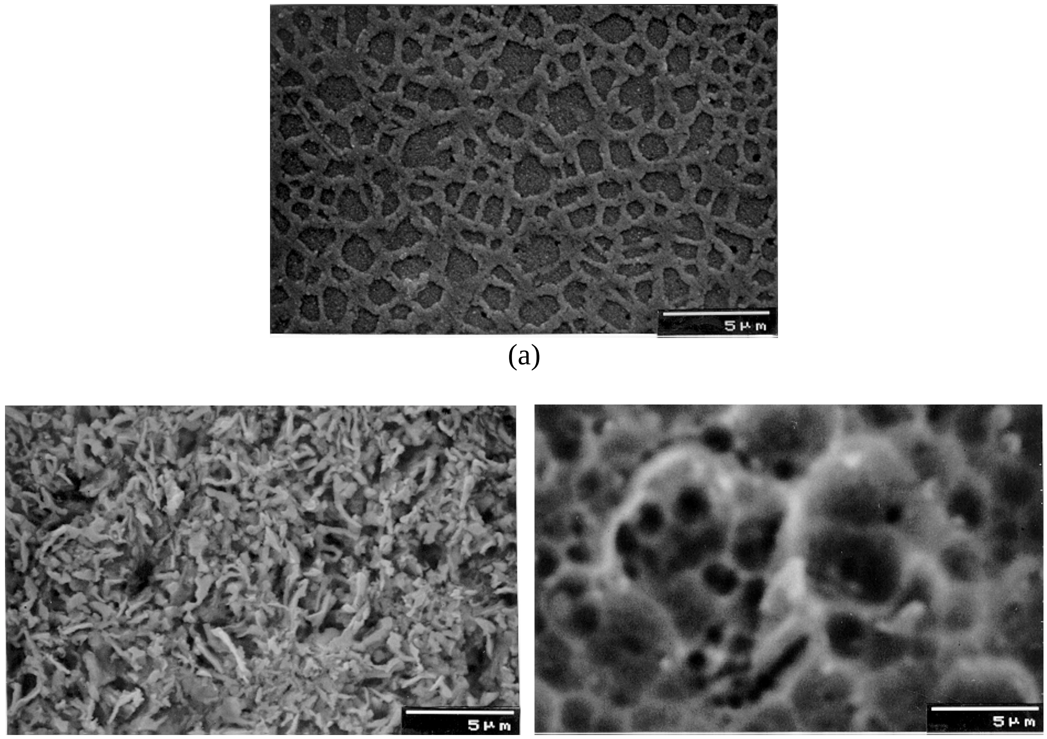


Fig. 1 Relationship between the Volume Fraction of U-10Mo Alloy and The Measured Density of the Powder Compacts.

Table 3 Description of U-Mo/Al Powder Compacts for Nano-plate

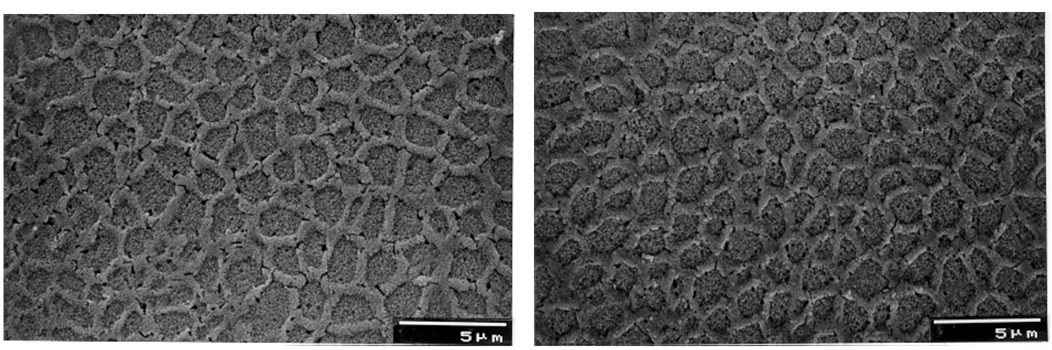
Fuel Composition	Compact I.D.	Alloy U-Density (g/cc)	Compact Density (g/cc)	Relative Density of Compact(%)	Volume Fraction of Fuel(%)
U-10Mo, As-atomized	Emo-372-001	15.32	9.722	0.955	0.507
	Emo-372-002	15.32	9.914	0.974	0.521
	Emo-372-003	15.32	9.826	0.965	0.514
	Emo-372-004	15.32	9.882	0.971	0.518
	Emo-372-005	15.32	10.123	0.994	0.536
	Emo-372-006	15.32	10.159	0.998	0.538
	Emo-372-007	15.32	9.938	0.976	0.522
U-10Mo, alpha +gamma	Emo-372-HT1-001	15.32	9.747	0.957	0.505
	Emo-372-HT1-002	15.32	10.076	0.990	0.528
	Emo-372-HT1-003	15.32	10.080	0.990	0.528
	Emo-372-HT1-004	15.32	10.128	0.995	0.532
	Emo-372-HT1-005	15.32	9.939	0.976	0.518
U-10Mo, Homo-genized	Emo-372-HT2-001	15.32	9.937	0.976	0.520
	Emo-372-HT2-002	15.32	10.162	0.998	0.536
	Emo-372-HT2-003	15.32	9.967	0.979	0.522
	Emo-372-HT2-004	15.32	9.877	0.970	0.516
	Emo-372-HT2-005	15.32	9.894	0.972	0.517
U-7Mo	Emo-378-001	16.31	9.572	0.960	0.490
	Emo-378-002	16.31	9.548	0.958	0.489
	Emo-378-003	16.31	9.560	0.959	0.489
	Emo-378-004	16.31	9.627	0.966	0.494
	Emo-378-005	16.31	9.689	0.972	0.499
	Emo-378-006	16.31	9.528	0.956	0.487
	Emo-378-007	16.31	9.552	0.958	0.489
	Emo-378-008	16.31	9.591	0.962	0.492
	Emo-378-009	16.31	9.449	0.948	0.482
U-6Mo	Emo-377-001	16.65	9.493	0.958	0.479
	Emo-377-002	16.65	9.654	0.974	0.490
	Emo-377-003	16.65	9.469	0.955	0.477
	Emo-377-004	16.65	9.452	0.954	0.476
	Emo-377-005	16.65	9.447	0.953	0.476
	Emo-377-006	16.65	9.586	0.967	0.485
	Emo-377-007	16.65	9.539	0.963	0.482
U-6.1Mo-0.9Ru	Emx-380-001	16.35	9.794	0.981	0.492
	Emx-380-002	16.35	9.736	0.976	0.488
	Emx-380-003	16.35	9.461	0.948	0.469
	Emx-380-004	16.35	9.794	0.981	0.492
	Emx-380-005	16.35	9.695	0.971	0.485
	Emx-380-006	16.35	9.772	0.979	0.491
	Emx-380-007	16.35	9.882	0.990	0.498
	Emx-380-008	16.35	9.740	0.976	0.489
	Emx-380-009	16.35	9.895	0.991	0.499
U-6Mo-1.7Os	Emx-381-001	16.42	9.886	0.984	0.494
	Emx-381-002	16.42	9.581	0.953	0.474
	Emx-381-003	16.42	9.924	0.987	0.497
	Emx-381-004	16.42	9.554	0.951	0.472
	Emx-381-005	16.42	9.826	0.978	0.490
	Emx-381-006	16.42	9.868	0.982	0.493
	Emx-381-007	16.42	9.825	0.978	0.490



(a)

(b) (c)

Fig. 2 Cross-sectional SEM Micrographs of As-atomized U-10Mo Powder, Decomposed U-10Mo Powder and Homogenized U-10Mo Powder; (a) As-atomized U-10Mo Powder, (b) Decomposed U-10Mo Powder, (c) Homogenized U-10Mo Powder.



(a) (b)

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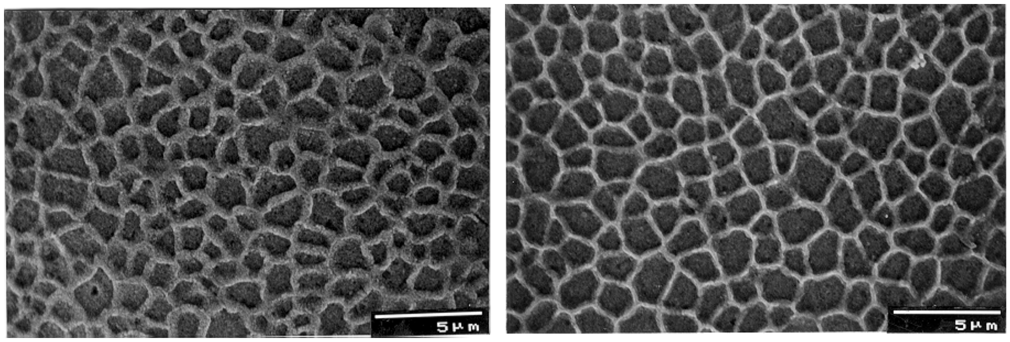


(c) (d)

Fig. 3 Cross-sectional SEM Micrographs of As-atomized U-6Mo Powder, U-7Mo Powder, U-6.1Mo-0.9Ru Powder and U-6.05Mo-1.7Os Powder; (a) U-6Mo Powder, (b) U-7Mo Powder, (c) U-6.1Mo-0.9Ru Powder, (d) U-6.05Mo-1.7Os Powder.

Table 4 Fabrication Conditions of Powder Compacts for Green Strength Measurement.

Number of Powder Compact	Fuel Powder	Volume Fraction of Fuel(%)
1	Atomized U-10Mo	40
2	Atomized U-10Mo	50
3	Atomized U-10Mo	60
4	Atomized U_3Si_2	50
5	Comminuted U_3Si_2	50

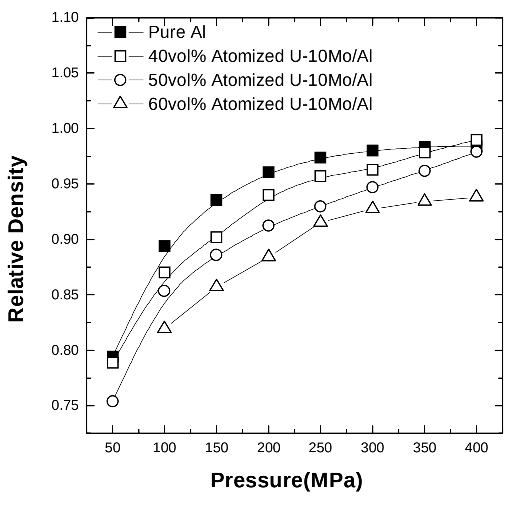


Fig. 4 The Relative Density of the Compacts of Pure Al, 40 vol%, 50 vol% and 60 vol% Atomized U-10Mo with Varying Pressure.

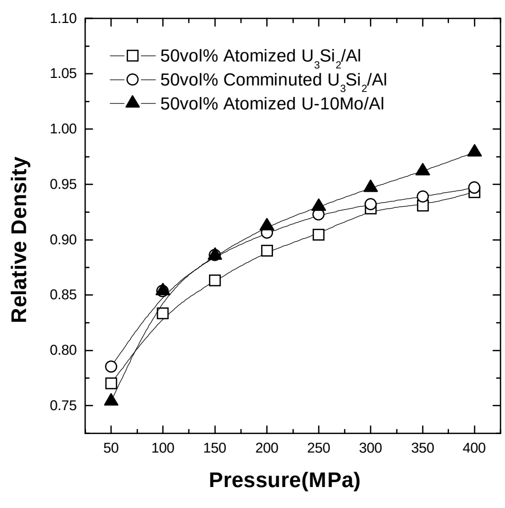
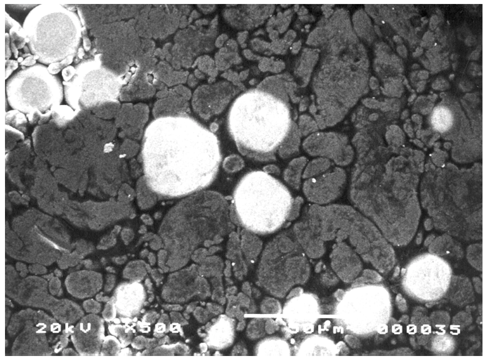
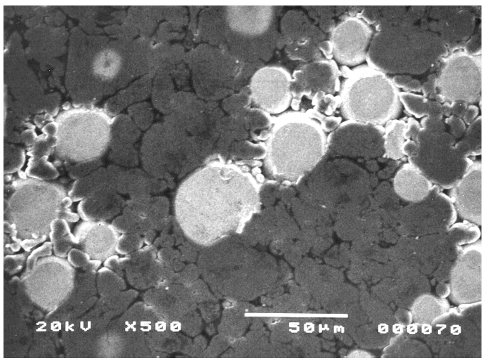


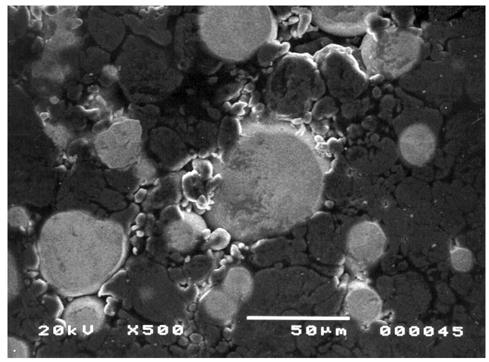
Fig. 5 The Relative Density of the Compacts of 50 vol% Atomized U-10Mo, 50 vol% Atomized U_3Si_2 and 50 vol% Comminuted U_3Si_2 with Varying Pressure.



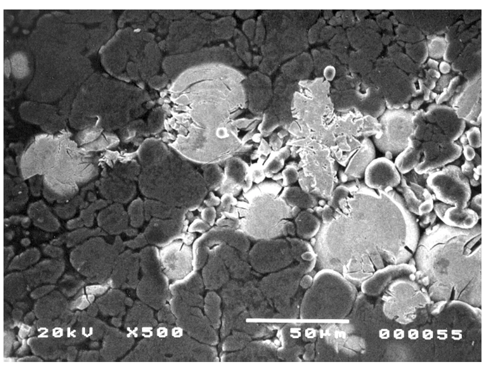
(a)



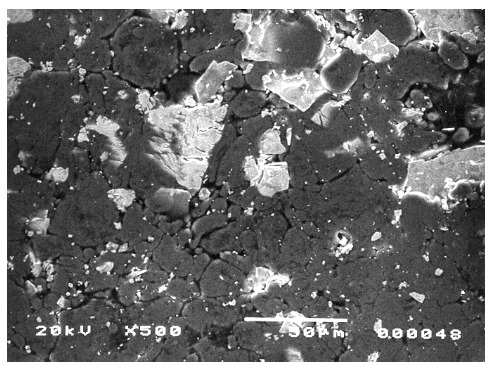
(b)



(c)



(d)



(e)

Fig. 6 SEM Micrographs of Powder Compacts Pressed at Pressure of 400 MPa; (a) 40 vol% Atomized U-10Mo/Al, (b) 50 vol% Atomized U-10Mo/Al, (c) 60 vol% Atomized U-10Mo/Al, (d) 50 vol% Atomized U_3Si_2/Al, (e) 50 vol% comminuted U_3Si_2/Al.

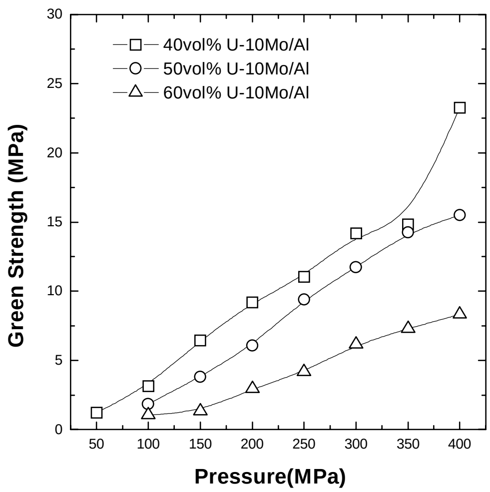


Fig. 7 The Green Strength of the Compacts of 40 vol%, 50 vol% and 60 vol% Atomized U-10Mo with Varying Pressures.

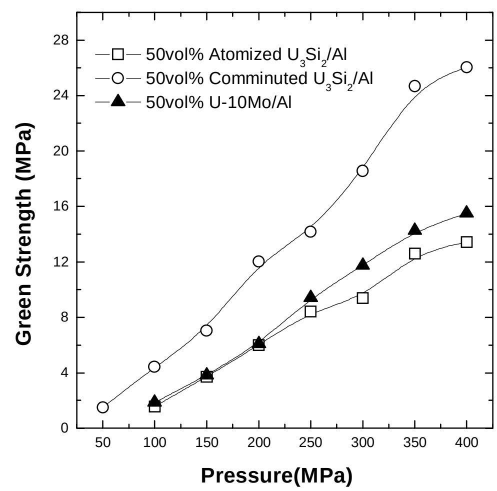


Fig. 8 The Green Strength of the Compacts of 50 vol% Atomized U-10Mo/Al, 50 vol% Atomized U_3Si_2 and 50 vol% Comminuted U_3Si_2 with Varying Pressures.

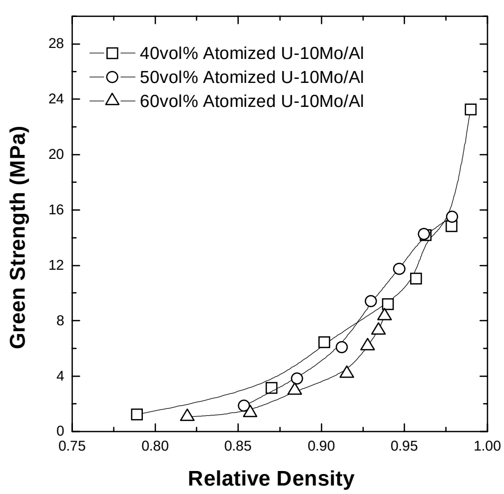


Fig. 9 The Green Strength of the Compacts of 40 vol%, 50 vol% and 60 vol% Atomized U-10Mo with Varying Relative Density.

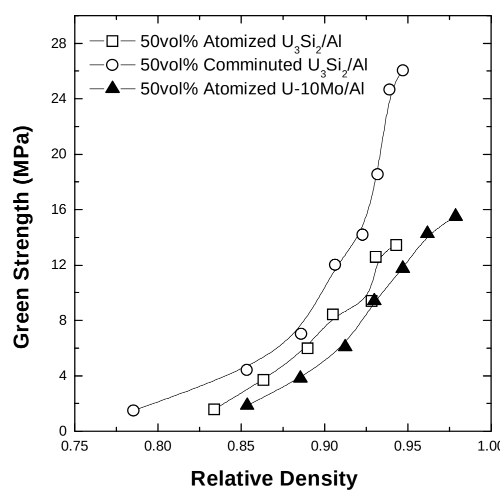


Fig. 10 The Green Strength of the Compacts of 50 vol% Atomized U-10Mo, 50 vol% Atomized U_3Si_2 and 50 vol% Comminuted U_3Si_2 with Varying Relative Density.